

Methanol as a tracer of fundamental constants

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ABSTRACT

The methanol molecule CH_3OH has a complex microwave spectrum with a large number of very strong lines. This spectrum includes purely rotational transitions as well as transitions with contributions of the internal degree of freedom associated with the hindered rotation of the OH group. The latter takes place due to the tunneling of hydrogen through the potential barriers between three equivalent potential minima. Such transitions are highly sensitive to changes in the electron-to-proton mass ratio, $\mu = m_e/m_p$, and have different responses to μ -variations. The highest sensitivity is found for the mixed rotation-tunneling transitions at low frequencies. Observing methanol lines provides more stringent limits on the hypothetical variation of μ than ammonia observation with the same velocity resolution. We show that the best quality radio astronomical data on methanol maser lines constrain the variability of μ in the Milky Way at the level of $|\Delta\mu/\mu| < 28 \times 10^{-9}$ (1σ) which is in line with the previously obtained ammonia result, $|\Delta\mu/\mu| < 29 \times 10^{-9}$ (1σ). This estimate can be further improved if the rest frequencies of the CH_3OH microwave lines will be measured more accurately.

Subject headings: molecular data — techniques: radial velocities — ISM: molecules — dark energy — elementary particles

1. Introduction

The hypothetical variation of the dimensionless physical constant μ – the electron-to-proton mass ratio – can be probed through the spectral observations of certain molecular transitions which are particularly sensitive to changes in μ . The corresponding sensitivity coefficients Q_μ of different molecular transitions relevant to astrophysics were calculated at first for H_2 (Varshalovich & Levshakov 1993), and later on for OH (Darling

2003; Chengalur & Kanekar 2003), $^{15}\text{ND}_3$ (van Veldhoven et al. 2004), H_2 (Ubachs et al. 2007), NH_3 (Flambaum & Kozlov 2007), OH and CH (Kozlov 2009), NH_2D and ND_2H (Kozlov et al. 2010), H_3O^+ (Kozlov & Levshakov 2011), CH_3OH (Jansen et al. 2011), and H_3O^+ , H_2DO^+ , HD_2O^+ , and D_3O^+ (Kozlov et al. 2011). Among them ammonia, NH_3 , is actively used in extragalactic and galactic observations of dense molecular clouds. In extragalactic observations the most stringent lim-

its on the mass ratio, $\Delta\mu/\mu = (\mu_{\text{space}} - \mu_{\text{lab}})/\mu_{\text{lab}}$, were obtained by Henkel et al. (2009) at redshift $z = 0.89$ and by Kanekar (2011) at $z = 0.69$: $|\Delta\mu/\mu| < 1400$ ppb (3σ), and $|\Delta\mu/\mu| < 400$ ppb (3σ), respectively (1ppb = 10^{-9}). Observations in the Milky Way have shown, however, a tentative signal $\Delta\mu/\mu = 26 \pm 3$ ppb (Levshakov et al. 2010a,b), but its nature remains unclear. To distinguish whether this is a real signal or an artefact due to unaccounted systematic effects, additional independent measurements involving other molecules are required.

One of such molecules — CH_3OH — was recently suggested by Jansen et al. (2011, hereafter J11). CH_3OH is a widespread interstellar molecule observed in the Milky Way, external galaxies (Herbst & van Dishoeck 2009; Sjouwerman et al. 2010; Martín et al. 2006), and even in comets (Bockelée-Morvan et al. 1991). The purpose of the present paper is to probe the variability of μ in the Milky Way using narrow emission lines of the methanol masers.

The microwave spectrum of CH_3OH is very rich because of the internal rotation of the OH group. The basic theory of the non-rigid tops with internal rotation was established in the fifties (Lin & Swalen 1959; Herschbach 1959) and the main features of the methanol spectrum were explained. Later on the theory was refined many times and currently there is a very impressive agreement between the theory and experiment (Anderson et al. 1990; Müller et al. 2006; Xu et al. 2008; Kleiner 2010).

2. Effective Hamiltonian and sensitivity coefficients

In this section the sensitivity coefficients Q_μ of the methanol lines are calculated. Our computational procedure differs from that described in J11. This allows us to check the values of Q_μ and their validity.

To calculate the microwave spectrum of CH_3OH we use an approach from Rabli & Flower (2010, hereafter RF), where a simple and convenient form of the effective Hamiltonian with six spectroscopic constants is suggested. This Hamiltonian is physically transparent and sufficiently accurate for calculations of sensitivity coefficients. All six parameters of the RF model have clear physical meaning

and their dependence on μ is easily understood within the Born-Oppenheimer approximation.

To show how rotational parameters scale with μ , we consider an example of a diatomic molecule in its ground vibrational state:

$$B_0 = \frac{1}{M} \left\langle v=0 \left| \frac{1}{R^2} \right| v=0 \right\rangle = B_e - \alpha_e + \dots, \quad (1)$$

where $B_e = 1/(MR_0^2)$ corresponds to the equilibrium internuclear distance R_0 and α_e is the vibrational correction. It is clear that $B_e \sim \mu$, but α_e has an additional dependence on μ via the vibrational wave function and, hence, scales as $\mu^{3/2}$. Thus, there is no one-to-one correspondence between terms of the effective Hamiltonian and the terms of the Born-Oppenheimer perturbation theory. As a result, B_0 scales as $\delta B_0/B_0 = Q_\mu^r \delta\mu/\mu$, with $Q_\mu^r \approx 1$. Typically, α_e/B_0 is of the order of 10^{-2} and Q_μ^r varies from 0.995 for the NO molecule to 0.981 for the H-bearing HF molecule.

We conclude that, in general, the rotational parameters A , B , and C scale linearly with μ within the uncertainty interval of 1–2%. These vibrational corrections are of the same order of magnitude as centrifugal corrections considered in J11. Both types of corrections are included below in the estimate of the error of the calculated sensitivity coefficients.

The rotational part of the Hamiltonian H_{rot} corresponds to the slightly asymmetric top and includes rotational constants A , B , and C ($B \approx C$). Here we use the standard convention $A > B > C$, while in RF $C > B > A$. The hindered rotation is described by the Hamiltonian

$$H_{\text{hr}} = -F \frac{d^2}{d\omega^2} + \frac{V_3}{2} (1 - \cos 3\omega), \quad (2)$$

where the kinetic coefficient F is proportional to μ and the electronic potential V_3 is independent on μ . This model does not include centrifugal distortions. Interaction of internal rotation with overall rotation is described by a single parameter D , which scales linearly with μ (Lin & Swalen 1959). For the rotational degrees of freedom we use the basis set of the prolate symmetric top and plane waves $\exp(im\omega)$ for the internal rotation. The Hamiltonian (2) mixes waves with $m' - m = 3n$. All relevant matrix elements are tabulated in RF. The fitted values of the parameters are also given there.

After the effective Hamiltonian is formed it is diagonalized numerically. Due to the C_3 symmetry of the Hamiltonian (2) the final eigenstates can be classified as A-type states and twofold degenerate E-type states. A-type states have definite parity p : for A^+ states $p = (-1)^J$ and for A^- states $p = (-1)^{J+1}$. Because methanol is close to the symmetric top its states are classified with an approximate quantum number K , which corresponds to the projection of the angular momentum \mathbf{J} on the axis of the CH_3 group. In this study we are interested only in the lowest states of the internal rotation of A and E symmetry: excitation of higher states requires kinetic temperatures $T \gtrsim 300\text{K}$, while warm molecular clouds have typical temperatures $T \lesssim 100\text{K}$.

The CH_3OH transitions with $\Delta K = 0$ can be considered approximately as rotational, where the state of the internal motion does not change. These transitions have ‘normal’ sensitivities to μ -variation with $Q_\mu \approx 1$. Because our model does not include centrifugal corrections, Q_μ must be exactly equal to 1. We estimate that these corrections do not exceed 3% for $J \leq 10$. The J11 model accounts for these corrections and provides the sensitivity coefficients for the $\Delta K = 0$ transitions between 1.00 and 1.03 in agreement with our estimates. We note that this deviation from unity is of the same order of magnitude as the vibrational correction which is not accounted for in both the RF and J11 effective Hamiltonians. The transitions with $\Delta K = \pm 1$, on the contrary, lead to the change in the internal motion and have sensitivities to μ -variation which vary in a wide range. Such transitions are of primary interest for our purpose.

In order to determine the sensitivity coefficients Q_μ for the microwave transitions we first find the dependence of the eigenvalues E_i on $\Delta\mu/\mu$:

$$\Delta E_i = q_i \frac{\Delta\mu}{\mu}, \quad (3)$$

where the coefficient q_i , individual for each level, shows a response of the level E_i to a small change of μ ($|\Delta\mu/\mu| \ll 1$). This is done by diagonalizing the effective Hamiltonian for three sets of the parameters, which correspond to $\mu = \mu_0$, and $\mu = \mu_0(1 \pm \varepsilon)$, where ε equals to 0.001 or 0.0001 (in both cases we obtain the same q -factors). The parameters A , B , C , D , and F , all scaling linearly

with μ , should then take the values A_0 , $A_0(1 \pm \varepsilon)$, etc., where the first value A_0 corresponds to the fit of the experimental spectrum. The dimensionless sensitivity coefficient Q_μ for the transition $\omega = E_{\text{up}} - E_{\text{low}}$ is found through calculation of the corresponding q -factor

$$\Delta\omega = q \frac{\Delta\mu}{\mu}, \quad (4)$$

where $q = q_{\text{up}} - q_{\text{low}}$, and

$$Q_\mu = \frac{q}{\omega}. \quad (5)$$

The results of calculations for transitions with $\Delta K = \pm 1$ are listed in Table 1, where we also give experimental and calculated transition frequencies. One can see that the accuracy of our model is about 1 GHz for all considered transitions. Consequently, the relative error for the high frequency transitions is only a fraction of a percent, but for the lowest frequency 6.7 GHz it is 15%. In order to improve the accuracy of the Q_μ -values we use experimental frequencies in Eq. (5). To check the computational procedure we also determined Q_μ for a few transitions with $\Delta K = 0$ ($0_0 - 1_0 A^+$ 48.4 GHz, $1_0 - 2_0 A^+$ 96.9 GHz, $1_1 - 2_1 A^+$ 96.1 GHz, $1_1 - 2_1 A^-$ 97.7 GHz, and $4_2 - 5_2 A^-$ 242.2 GHz) and for all of them found $Q_\mu = 1.00$.

Table 1 also lists the errors of the sensitivity coefficients in their last digits which are given in parenthesis. These errors were estimated in the following way. The effective Hamiltonian from RF includes the rotational and tunneling parts as well as the interaction between them. The transition frequencies and the sensitivity coefficients are found from the numerical diagonalization of this Hamiltonian followed by the numerical differentiation of the results in respect to μ .

Only the low frequency transitions can have an enhanced sensitivity to the μ -variation because they are of a mixed character with the rotational and tunneling contributions to the transition energy:

$$\omega = \omega_r - \omega_t. \quad (6)$$

These two contributions have the following dependences on μ :

$$\frac{\Delta\omega_r}{\omega_r} = Q_\mu^r \frac{\Delta\mu}{\mu}, \quad \frac{\Delta\omega_t}{\omega_t} = Q_\mu^t \frac{\Delta\mu}{\mu}. \quad (7)$$

Then, the resultant sensitivity of the transition ω is given by:

$$\frac{\Delta\omega}{\omega} = \left(Q_\mu^r \frac{\omega_r}{\omega} - Q_\mu^t \frac{\omega_t}{\omega} \right) \frac{\Delta\mu}{\mu} \equiv Q_\mu \frac{\Delta\mu}{\mu}. \quad (8)$$

Let us suppose that the sensitivities Q_μ^r and Q_μ^t are known with a relative error ε . Then, the absolute error of Q_μ is equal to:

$$\Delta Q_\mu = \varepsilon \times \frac{Q_\mu^r \omega_r + Q_\mu^t \omega_t}{\omega}. \quad (9)$$

In order to use this expression to estimate the errors ΔQ_μ we need to know the decomposition (6) and the sensitivity coefficients Q_μ^r and Q_μ^t . As we discussed earlier, $Q_\mu^r \approx 1$. We can estimate the tunneling sensitivity to be $Q_\mu^t \approx 2.6$ from the semi-classical (WKB) approximation (e.g., Kozlov et al. 2010). The tunneling part of the transition energy ω_t can be estimated from the model of Hecht & Dennison (1957). The rotational energy ω_r is then given by the experimental frequency ω and Eq. (6).

We conservatively estimate the relative error of our calculations of Q_μ^r and Q_μ^t to be of about 3%, i.e., $\varepsilon = 0.03$. This error is associated with the missing centrifugal distortion and vibration correction which changes the rotational and tunneling frequencies by a few percent at the most. The resulting errors ΔQ_μ are estimated from Eq. (9).

Table 1 shows that the sensitivity coefficients of the mixed rotation-tunneling transitions at low frequencies lie in the range between $-17(1)$ and $+43(3)$. For comparison, the H_2 Lyman and Werner transitions have $Q_\mu \sim 0.03$ (Varshalovich & Levshakov 1993), and the ammonia inversion transition has $Q_\mu = 4.46$ (Flambaum & Kozlov 2007). This means that methanol is almost 1000 times more sensitive to the change in μ than molecular hydrogen and about 10 times more sensitive than ammonia. Besides, CH_3OH has many lines with different sensitivities of both signs which allows us to estimate $\Delta\mu/\mu$ from observations of only this one molecule. This is the advantage over, e.g., ammonia NH_3 which has the same sensitivity coefficients for the inversion transitions (1,1), (2,2), etc., and, hence, requires some rotational transitions of other molecule as a reference in order to trace $\Delta\mu/\mu$.

Now we can compare our Q_μ values with the sensitivity coefficients K_μ derived in J11. We note

that μ was defined in J11 as m_p/m_e and, hence, one expects $Q_\mu = -K_\mu$. This comparison is shown graphically in Fig. 1. The error bars in this figure mark the 1σ uncertainties. In total we have 35 common computations of the sensitivity coefficients. The theoretical calculations of their values show a good concordance except for the six points at 9.936, 37.703, 38.293, 38.453, 86.615, and 86.902 GHz where the discrepancy $|Q_\mu + K_\mu|$ exceeds the 2σ level (in Fig. 1, the corresponding Q_μ values are -14, 5.1, 12.1, 12.1, 5.9, and 5.9). The reason for such deviations is not clear. The highest offsets of 2.8σ and 2.7σ are found for the $J = 5-6$ transitions at 38.293 and 38.452 GHz, respectively, where the RF model should have a sufficiently accurate result. On the other hand, the errors of the K_μ values were taken in J11 to be 5% if $|K_\mu| \geq 1$ (relative error) or 0.05 if $|K_\mu| < 1$ (absolute error). However, analysis based on Eq. (9) shows that the relative errors noticeably differ from line to line.

3. Observational constraints on $\Delta\mu/\mu$

The agreement between the values determined with independent methods gives us confidence that, given the high Q_μ values with different signs, methanol is excellent for testing the electron-to-proton mass ratio. Below we consider such a test based on high angular and high spectral resolution observations of CH_3OH maser lines.

Equations (4) and (5) show that for a given transition from Table 1, ω_i , with the sensitivity coefficient Q_i , the expected frequency shift, $\Delta\omega_i/\omega_i$, due to a change in μ is given by

$$\frac{\Delta\omega_i}{\omega_i} = Q_i \frac{\Delta\mu}{\mu}. \quad (10)$$

Then the value of $\Delta\mu/\mu$ can be estimated from two transitions with different sensitivity coefficients Q_i and Q_j :

$$\frac{\Delta\mu}{\mu} = \frac{V_j - V_i}{c(Q_i - Q_j)}, \quad (11)$$

where V_j and V_i are the apparent radial velocities of the corresponding CH_3OH transitions and c is the speed of light.

Interstellar CH_3OH lines were widely observed in the last two decades and we can obtain some preliminary estimates of $\Delta\mu/\mu$ using the published data. In particular, the maser CH_3OH emission is

Table 1: Numerical calculation of the Q -factors for the low frequency mixed rotation-tunneling transitions ($\Delta K = \pm 1$) in methanol. The rest frequencies are taken from Müller et al. (2004)[†] except for those marked by the asterisk which are from Lovas (2004). The 1σ uncertainties of the least significant figure of the rest frequencies and Q_μ -values are given in parenthesis.

Transition $J_{lK_l} - J_{uK_u}$	ω (MHz)		Q_μ	Transition $J_{lK_l} - J_{uK_u}$	ω (MHz)		Q_μ
	Exper.	Theor.			Exper.	Theor.	
$6_0-5_1 A^+$	6668.5192(4)	5777.8	+43(3)	$4_0-5_{-1} E$	84521.169(10)	85413.3	-3.5(4)
$8_{-2}-9_{-1} E$	9936.202(2)	11281.9	-14(1)	$6_3-7_2 A^-$	86615.600(5)	86033.5	+5.9(3)
$3_{-1}-2_0 E$	12178.597(2)	11487.4	+32(2)	$6_3-7_2 A^+$	86902.949(5)	86315.2	+5.9(3)
$3_0-2_1 E$	19967.3961(2)	20012.6	+6.3(3)	$7_1-8_0 A^+$	95169.463(10)	96244.8	-1.9(3)
$3_1-3_2 E$	24928.707(7)	25565.7	-17(1)	$10_{-2}-11_{-1} E$	104300.414(7)	105949.0	-0.45(16)
$4_1-4_2 E$	24933.468(2)	25597.1	-17(1)	$4_0-3_1 A^+$	107013.803(5)	106283.3	+3.6(2)
$2_1-2_2 E$	24934.382(5)	25551.1	-17(1)	$1_{-1}-0_0 E$	108893.963(7)	108370.8	+4.5(2)
$5_1-5_2 E$	24959.0789(4)	25655.3	-17(1)	$5_0-6_{-1} E$	132890.692(10)	133898.0	-1.9(3)
$6_1-6_2 E$	25018.1225(4)	25752.6	-16(1)	$8_1-9_0 A^+$	146618.794(50)	147799.4	-0.9(2)
$7_1-7_2 E$	25124.8719(4)	25902.7	-16(1)	$8_{-1}-8_0 E$	156488.868(10)	155888.7	+3.4(2)
$8_1-8_2 E$	25294.4165(2)	26120.3	-16(1)	$3_0-2_1 A^+$	156602.413(10)	155946.7	+2.8(1)
$9_1-9_2 E$	25541.3979(4)	26419.4	-16(1)	$7_{-1}-7_0 E$	156828.533(10)	156258.3	+3.4(2)
$3_1-4_0 E$	28316.031(8)*	28351.5	-2.8(3)	$6_{-1}-6_0 E$	157048.625(10)	156506.3	+3.4(2)
$9_1-8_2 A^-$	28969.942(50)	29091.1	+11.1(6)	$5_{-1}-5_0 E$	157179.017(10)	156661.9	+3.4(2)
$3_0-4_{-1} E$	36169.265(30)	36956.3	-9.6(9)	$4_{-1}-4_0 E$	157246.056(10)	156750.9	+3.4(2)
$8_{-1}-7_{-2} E$	37703.700(30)	36486.0	+5.1(3)	$1_{-1}-1_0 E$	157270.851(10)	156817.2	+3.4(2)
$5_3-6_2 A^-$	38293.268(50)	37660.7	+12.1(7)	$3_{-1}-3_0 E$	157272.369(10)	156795.3	+3.4(2)
$5_3-6_2 A^+$	38452.677(50)	37817.3	+12.1(7)	$2_0-2_1 A^+$	304208.324(13)*	303751.3	+1.91(7)
$6_1-7_0 A^+$	44069.410(10)	45048.7	-5.3(6)	$4_0-4_1 A^+$	307165.911(13)*	306658.4	+1.89(7)

Note. — [†]The uncertainties of the first three transitions at 6.668, 9.936, and 12.178 GHz given in Müller et al. (2004) correspond to the 2σ errors in accord with Breckenridge & Kukolich (1995).

of a special interest here since maser lines are narrow and the error of the line center measurement is correspondingly low.

The CH_3OH molecules originate in star-forming regions and are observed as maser emission in two types of sources: class I and class II (e.g., Menten 1991). Class II methanol masers are radiatively pumped and located in the vicinity of young stellar objects (YSOs), whereas class I methanol masers are believed to trace distant parts of the outflows from YSOs and are collisionally pumped. The most accurate $\Delta\mu/\mu$ values can be estimated from narrow and symmetric line profiles with similar shapes. Such profiles are provided in interferometric observations of the class I masers located in the vicinity of IRAS 16547-4247 (G343.12-0.06) which is a luminous YSO with a radio jet (Voronkov et al. 2006, hereafter V06). This methanol maser emission consists of a clus-

ter of six spots spread over an area of 30 arcsec in extent. One spot (called *B* in V06) shows activity in 12 CH_3OH transitions (Fig. 3 of V06). Among them, the 9.9 GHz and 104 GHz lines have a narrow spike ($FWHM < 30 \text{ m s}^{-1}$) on top of a broader ($FWHM \approx 300 \text{ m s}^{-1}$) symmetric line. The widths of these spikes do not exceed the channel spacing which is 29 m s^{-1} and 22 m s^{-1} at 9.9 GHz and 104 GHz, respectively. According to V06, these are the narrowest spectral features ever found. Their linewidths imply that the masers are unsaturated and that the turbulent motion in the gas is strongly suppressed. The 9.9 GHz and 104 GHz masers show similar angular sizes, $\theta_{9.9} = 0.10 \pm 0.09 \text{ arcsec}$, and $\theta_{104} = 0.2 \pm 0.1 \text{ arcsec}$, and almost the same coordinates, $|\Delta\alpha| = 0.03 \pm 0.04 \text{ arcsec}$, and $|\Delta\delta| = 0.09 \pm 0.04 \text{ arcsec}$. The lines were observed in a month interval on June 16, 2005 (9.9 GHz)

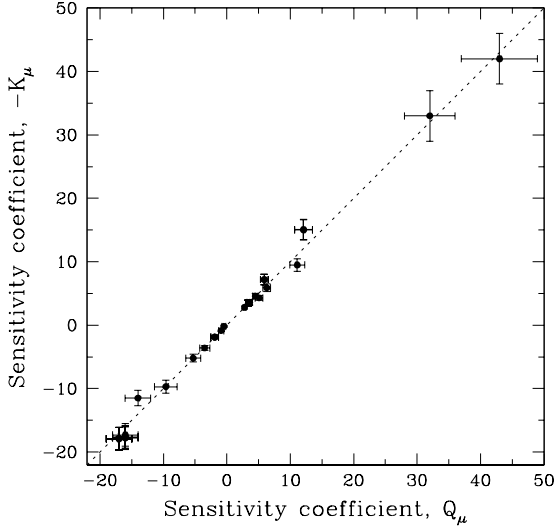


Fig. 1.— Comparison of the sensitivity coefficients calculated in the present paper (Q_μ) with those from J11 ($-K_\mu$). Shown by error bars are the 2σ uncertainties.

and on Aug 18, 2005 (104 GHz).

As noted in V06, the 9.9 GHz and 104 GHz transitions belong to the same $J_{-2} - (J+1)_{-1}$ E transition series with $J = 8$ and 10, respectively, and are expected to show a similar behavior, i.e., both maser lines should originate in the same volume and, hence, have equal radial velocities. The radial velocities of these spikes, $V_{9.9} = -31.554$ km s $^{-1}$ and $V_{104} = -31.594$ km s $^{-1}$, are measured with the uncertainty of a few m s $^{-1}$, but, unfortunately, their difference, $\Delta V = V_{9.9} - V_{104} = 40$ m s $^{-1}$, is less certain because of the errors in the rest frequencies, $\varepsilon_{9.9} = 60$ m s $^{-1}$ and $\varepsilon_{104} = 20$ m s $^{-1}$ (see Table 1). Since at these two frequencies the sensitivity coefficients are slightly different for two calculations [$Q_{\mu,9.9} = -14(1)$, $-K_{\mu,9.9} = -11.5(6)$, and $Q_{\mu,104} = -0.45(16)$, $-K_{\mu,104} = -0.18(5)$], we use their average values $\bar{Q} = (Q_\mu - K_\mu)/2$ to estimate $\Delta\mu/\mu$ from Eq. 11. Thus, with $\bar{Q}_{\mu,9.9} = -12.75(58)$, $\bar{Q}_{\mu,104} = -0.32(8)$ and $\Delta V = 40(63)$ m s $^{-1}$ we find $\Delta\mu/\mu = 11 \pm 17$ ppb (1σ , c.l.) or the upper limit $|\Delta\mu/\mu| < 28$ ppb. This single point estimate is in line with a limit derived from the sample mean of ammonia observations in the Milky Way,

$\Delta\mu/\mu = 26 \pm 3$ ppb (1σ , c.l.), or $|\Delta\mu/\mu| < 29$ ppb (Levshakov et al. 2010a).

The reliability of the present estimate of $\Delta\mu/\mu$ can be further improved if new laboratory frequencies of the methanol 9.9 GHz and 104 GHz transitions will be determined with a higher accuracy. Since maser sources are in general variable in time, additional gains can be obtained from simultaneous observations of several methanol lines in a way described, e.g., in Voronkov et al. (2011) where up to eight methanol maser transitions were observed simultaneously with the Australia Telescope Compact Array (ATCA). Unfortunately, the observed methanol profiles at 24, 25 GHz and 9.9 GHz have close sensitivity coefficients with $\Delta Q \approx 3$ which is comparable to the ammonia method. This does not allow us to improve the aforementioned upper limit on $\Delta\mu/\mu$. Without new laboratory studies and improvements in astronomical observations, which require substantial care to determine frequencies with an accuracy better than 10^{-8} , any further advances in exploring $\Delta\mu/\mu$ from methanol maser spectra will be impossible.

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